

PATENT  
Attorney Docket No. 07040.0224-00

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

National Stage of International Application )  
No. PCT/EP2002/014145 under 35 U.S.C. )  
§ 371 ) Group Art Unit: 1791  
Inventors: Maurizio GALIMBERTI et al. )  
Application No.: 10/534,545 ) Examiner: J. FISCHER  
§ 371 Date: October 27, 2005 ) Confirmation No.: 6976  
PCT filed: December 12, 2002 )  
For: TYRE FOR VEHICLE WHEELS AND )  
ELASTOMERIC COMPOSITION )

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

**DECLARATION UNDER 37 C.F.R. § 1.132**

I, Giuseppina Ratti, declare and state that:

1. I am an Italian, residing at Seregno (Milano), Via Verdi 127.
2. I have been awarded a degree in industrial chemistry from the University of Milan, and am a Doctor in industrial chemistry.
3. I have been employed by Pirelli S.p.A. ("Pirelli") since November 1987 and I am presently a tyre materials compounding technologist of the Pirelly Tyre at Pirelli. During my employment at Pirelli, I have been engaged in research and development regarding tyre materials.

4. Given my education and experience, particularly in the area of materials, I consider myself able to provide the following testimony based on experiments conducted under my direct supervision.

**TESTING**

5. A comparative elastomeric composition containing Duoquad® T/50 was compared with (1) the comparative elastomeric compositions of Examples 4-6 of present U.S. Patent Application No. 10/534,545 ("the '545 application) and (2) the inventive elastomeric compositions of Examples 7 and 8 of the '545 application. Instead of being a quaternary ammonium salt of the claimed invention, Duoquad® T/50 (commercialized by Lion Akzo Co., Ltd.) is a diquaternary ammonium chloride, i.e., an ammonium salt having two nitrogen atoms that are both quaternized (i.e., both have a positive charge). See attached Duoquad Product Data Sheet. Duoquad® T/50 appears to be structurally identical to Redicote E-11, the product reported in U.S. Patent No. 3,686,113 to Burke ("Burke"). See, e.g., Burke, col. 12 (Table VIII, footnote 1) (defining Redicote E-11 as  $C_{18}H_{37}N(CH_3)_2ClC_3H_6N(CH_3)_3Cl$ ).

6. The compositions of the comparative elastomeric composition containing Duoquad® T/50 (identified as Example 10), the comparative elastomeric compositions of Examples 4-8, and the inventive elastomeric compositions of Examples 7 and 8 are listed in Table A (amounts in PHR):

TABLE A

	EXAMPLE					
	4 <sup>(*)</sup>	5 <sup>(*)</sup>	6 <sup>(*)</sup>	7	8	10 <sup>(*)</sup>
<b>1<sup>st</sup> Step</b>						
S-SBR	90	90	90	90	90	90
BR	35	35	35	35	35	35
Silica	70	70	70	70	70	70
TESPT	5.6	5.6	5.6	5.6	5.6	5.6
Stearic acid	2	2	2	2	2	2
Aromatic oil	8	8	8	8	8	8
Microcrystalline wax	1	1	1	1	1	1
<b>2<sup>nd</sup> Step</b>						
Zinc oxide	2.5	2.5	2.5	2.5	2.5	2.5
Antioxidant	2	2	2	2	2	2
<b>3<sup>rd</sup> Step</b>						
Bardac® LF-80	-	-	2.8	-	-	-
Ammonium salt (1)	-	-	-	2.8	-	-
Ammonium salt (2)	-	-	-	-	2.8	-
Duoquad® T/50						2.8
DPG	1.9	-	-	-	-	-
CBS	2	2	2	2	2	2
Sulphur	1.2	1.2	1.2	1.2	1.2	1.2

(\*): comparative

With the exception of Duoquad® T/50, which is described above, each of the

ingredients identified in Table A are described in the '545 application.

### PROCEDURE

7. The comparative elastomeric compositions (Examples 4-6 and 10) and inventive elastomeric compositions (Examples 7 and 8) were prepared according to the procedure set forth in paragraphs [0126] and [0127] of the '545 application as-published (U.S. Patent Application Publication No. 2006/0155077 A1).

## RESULTS

8. Applying the testing procedures described in the '545 application, the comparative and inventive elastomeric compositions had the following physical characteristics, as shown in Table B:

TABLE B

	<b>EXAMPLE</b>					
	<b>4<sup>(*)</sup></b>	<b>5<sup>(*)</sup></b>	<b>6<sup>(*)</sup></b>	<b>7</b>	<b>8</b>	<b>10<sup>(*)</sup></b>
Mooney viscosity ML (1+4)	74.20	87.20	65.40	76.70	76.60	83.20
<b>STATIC MECHANICAL PROPERTIES</b>						
Stress at break (MPa)	14.11	16.24	13.41	14.97	13.91	14.27
Elongation at break (MPa)	426.00	612.10	406.90	440.50	374.30	466.20
<b>DYNAMIC MECHANICAL PROPERTIES</b>						
E' (23° C)	8.277	7.735	6.974	7.791	7.784	7.539
E' (70° C)	5.988	5.446	5.510	5.940	5.953	5.144
T <sub>delta</sub> (23° C)	0.268	0.282	0.224	0.233	0.226	0.262
T <sub>delta</sub> (70° C)	0.140	0.165	0.110	0.122	0.120	0.145
<b>RHEOMETRIC PROPERTIES</b>						
ML (dN M)	3.08	4.03	2.20	2.75	3.09	3.95
MH (dN m)	20.51	22.07	17.89	19.05	18.99	21.50
t <sub>30</sub> (min)	2.13	2.40	2.16	1.52	1.06	2.32
t <sub>90</sub> (min)	4.12	13.18	5.63	5.64	3.69	12.50
IRHD Hardness (23° C)	69.7	66.9	65.3	67.1	68.8	65.9
IRHD Hardness (100° C)	64.7	60.8	62.1	62.7	63.6	58.9
DIN Abrasion	86.3	74.3	100.7	85.1	84.4	77.5

(\*): comparative

## CONCLUSION

9. The results of this testing illustrate that comparative Example 10 containing Duoquad® T/50 (i.e., an ammonium salt according to Burke (i.e., Redicote

E-11) has a high value for its Mooney viscosity (i.e., poor processability of the rubber mixture), which is only slightly lower than comparative Example 5, which is devoid of DPG and any ammonium salt (i.e., a composition devoid of any secondary accelerators). In contrast, inventive Examples 7 and 8, which contained an ammonium salt according to the claims, provided a lower Mooney viscosity as compared to Examples 5 and 10 and substantially identical to comparative Example 4, which contained DPG, a secondary accelerator.

10. As to the static and dynamic mechanical properties, the data presented in Table B do not indicate any remarkable differences.

11. As to the MDR rheometric analysis, comparative Example 10 provided unsatisfactory results, since the parameters of the rheometric curve were substantially identical to those of comparative Example 5 (i.e., a composition devoid of any secondary accelerators (e.g., DPG or ammonium salt)). In particular, the inventive Examples 7 and 8 had values for t<sub>30</sub> (min) and t<sub>90</sub> (min) (i.e., the time required to reach 30% or 90% of the maximum torque M<sub>H</sub>) that are acceptable for an industrial application, and that substantially correspond to, or are even better than, those achieved by comparative Example 4, which only contained DPG.

12. Accordingly, the comparative tests demonstrate both (1) that the quaternary ammonium salts according to the claimed invention can be advantageously used as secondary accelerators in replacement of DPG in rubber compositions reinforced with silica. and (2) the diquaternary ammonium salts of Burke, such as Redicote E-11, are unsuitable for that very purpose, as evidenced by their respective

effects on the vulcanization rate (see the rheometric data) and on processability (see the Mooney viscosity values) of the rubber mixtures.

13. Based on my education and experience, it is my opinion that it would have been unexpected that elastomeric compositions containing the claimed ammonium salts would have significantly better physical characteristics in terms of vulcanization rate and processability as compared with elastomeric compositions containing ammonium salts outside the scope of the claims, such as Duoquad® T/50 (or Redicote E-11).

14. I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: 36 / 1 / 2009

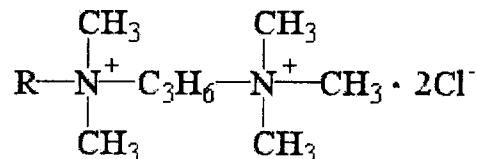
By: Giuseppina Ratti  
Giuseppina Ratti

1.TYPICAL PROPERTIES  
2.TYPICAL COMPOSITIONS  
3.PRODUCT INFORMATION

[Top of Quaternary ammonium salts]

**Product Classification:** **Quaternary ammonium salts**  
(Diquaternaryammonium chlorides)

**Product Name:** **Duoquad**



**1.TYPICAL PROPERTIES**

Product Name	Color(Gardner)	Ash(%)	pH	Appearance@25°C
Duoquad T/50	<10	<1	7.5	Liquid

**2.TYPICAL COMPOSITIONS**

Product Name	Chemical Name	Active Ingredient(%)	IPA* (%)	Moisture (%)
Duoquad T/50	Tallowalkylpentamethyl propylenediammonium dichloride	50	35	15

\* IPA:Isopropanol

**3.PRODUCT INFORMATION**

Product Name	CAS No.	ENCS No.*	Packing(Net: kg)	
			Drum	Can
Duoquad T/50	68607-29-4	7-6	-	-

\* ENCS:The list of Existing and New Chemical Substances promulgated by the Kashin Act of Japan.

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